# Geochemical Evolution of Highly Alkaline and Saline Tank Waste Plumes During Seepage Through Vadose Zone Sedments

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## RESEARCH OBJECTIVES

Leakage of highly saline and alkaline radioactive waste solutions from storage tanks into underlying sediments is a serious problem at the Hanford Site in Washington State. Although it was found from field samples that pH values of the initially highly alkaline (pH 14) waste plumes dramatically decreased (to pH 10–7), understanding of the neutralization process was lacking. Since pH is a master geochemical variable, the behavior of waste plume contaminants, including their speciation, sorption, solubility, precipitation, and transport, can be reliably predicted only when the evolution of the pH profile is understood. This study focuses on the geochemical evolution of major geochemical parameters including pH, and addresses how pH evolved as the plumes propagated.

## APPROACH

We designed a plume profiling method to obtain spatially and temporally direct measurements of plume geochemistry profiles. The influences of waste solution pH, ionic strength, and sediment type were also studied. To maximize the relevance of this laboratory study for understanding real field problems, most of the experiments involved infusion of synthetic waste solutions into sediment columns, thereby integrating the influences of reactions and transport.

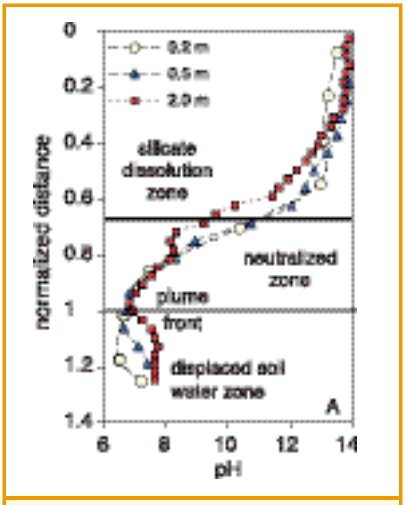


Figure 1. The pH profile along the 0.2, 0.5, and 2.0 m tall columns, with the distance axis normalized to the predicted plume front position

## ACCOMPLISHMENTS

This study revealed that while the plume is connected to an actively leaking source, its profile spans the very broad range from pH 14 (influent waste pH) within the near-source region, down to pH 6.5 (lower than that of the initial soil solution) at the plume front (Figure 1). The plume can be divided into two zones: the Silicate Dissolution Zone (SDZ, pH 14–10), and Neutralized Zone (NZ, pH 10–7). After the plume source became inactive and the plume aged, pH values within the SDZ continued to decrease at a decreasing rate and eventually reached equilibrium at around pH 10—whereas the pH values in the original neutralized zone remained relatively unchanged. The major reactions responsible for the pH evolution of the waste plumes were identified, along with specific regions within the plumes where they occur.

## SIGNIFICANCE OF FINDINGS

This laboratory study provides information on how pH evolved as tank waste plumes propagated. As a master geochemical variable, pH needs to be understood in order to predict the fate and transport of contaminants carried by waste plumes.

## RELATED PUBLICATIONS

Wan, J., T.K. Tokunaga, J.T. Larsen, and R.J. Serne, Geochemical evolution of highly alkaline and saline tank waste plumes during seepage through vadose zone sediments. Geochim. Cosmochim. Acta, 2003 (in press).

Wan, J., J.T. Larsen, T.K. Tokunaga, and Z. Zheng, pH neutralization and zonation in alkaline-saline tank waste plumes. Environ. Sci. Technol., 2003 (submitted).

## ACKNOWLEDGMENTS

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